Mapping the Generator Coordinate Method to the Coupled Cluster Approach

Jason L. Stuber*
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The generator coordinate method (GCM) casts the wavefunction as an integral over a weighted set of non-orthogonal single determinantal states. In principle this representation can be used like the configuration interaction (CI) or shell model to systematically improve the approximate wavefunction towards an exact solution. In practice applications have generally been limited to systems with less than three degrees of freedom. This bottleneck is directly linked to the exponential computational expense associated with the numerical projection of broken symmetry Hartree-Fock (HF) or Hartree-Fock-Bogoliubov (HFB) wavefunctions and to the use of a variational rather than a bi-variational expression for the energy. We circumvent these issues by choosing a hole-particle representation for the generator and applying algebraic symmetry projection, via the use of tensor operators and the invariant mean (operator average). The resulting GCM formulation can be mapped directly to the coupled cluster (CC) approach, leading to a significantly more efficient approach than the conventional GCM route.

I. THE GENERATOR COORDINATE METHOD (GCM)

The generator coordinate method (GCM), initially developed to treat large amplitude collective motion in nuclei [1–3] (see also Refs. [4, 5]), represents the wavefunction as an integral over a weighted set of states

$$|\Psi_{\rm GCM}\rangle = \int d\mathbf{Z}\, f(\mathbf{Z})\, |\Phi(\mathbf{Z})\rangle\,, \tag{1}$$

each state $|\Phi(\mathbf{Z})\rangle$ parameterized by a continuous variable $\mathbf{Z}$, the so-called generator coordinate. For bosonic modes this structural form is related to the coherent state representation [6]. Unlike the configuration interaction (CI) or shell model

$$|\Psi_{\rm CI}\rangle = \sum_{\mu} C_{\mu}\, |\Phi_{\mu}\rangle\,, \tag{2}$$

which expands the wavefunction as a linear combination of mutually orthogonal states

$$\langle\Phi_{\mu}|\Phi_{\nu}\rangle = \delta_{\mu\nu}\,, \tag{3}$$

the states $|\Phi(\mathbf{Z})\rangle$ which comprise the GCM are not mutually orthogonal by design, namely the state overlap matrix

$$\mathbf{N}(\mathbf{Z}';\mathbf{Z}) = \langle\Phi(\mathbf{Z}')|\Phi(\mathbf{Z})\rangle\,, \tag{4}$$

is not diagonal.

Of primary interest in this work are fermions (electrons or nucleons) for which each basis state $|\Phi(\mathbf{Z})\rangle$ is then a Slater determinant, weighted by the complex valued scalar function $f(\mathbf{Z})$ in Eq. (1). Thouless' Theorem [7, 8] parametrizes these determinants by an exponential acting on a reference state $|\Phi_0\rangle$

$$|\Phi(\mathbf{Z})\rangle = e^{\hat{\rm Z}}\, |\Phi_0\rangle\,, \tag{5}$$

where, schematically

$$\hat{\rm Z} = \sum_{\mu} Z_{\mu} \hat{\rm E}_{\mu}\,, \tag{6}$$

is a mono-excitation level generator with corresponding coefficients $\mathbf{Z} = (Z_{\mu})$, $\hat{\rm E}_{\mu}$ is the product of two fermion creation and/or annihilation operators. The structure of $\hat{\rm Z}$ and $|\Phi_0\rangle$ in turn define the space spanned by all possible GCM wavefunctions. If the generators $\hat{\rm Z}$ are not sufficiently general, breaking one or more symmetries of the hamiltonian, then it is not possible to represent all symmetry preserving wavefunctions.

Solutions for the GCM wavefunction are obtained in the time-independent case via the Schrödinger equation

$$\hat{\rm H}\, |\Psi_{\rm GCM}\rangle = E\, |\Psi_{\rm GCM}\rangle\,, \tag{7}$$

which can be solved via a matrix representation, resulting in the Hill-Wheeler-Griffin (HWG) equation

$$\int d\mathbf{Z}\, \mathbf{H}(\mathbf{Z}';\mathbf{Z})\, f(\mathbf{Z}) = E \int d\mathbf{Z}\, \mathbf{N}(\mathbf{Z}';\mathbf{Z})\, f(\mathbf{Z})\,, \tag{8}$$

which is a generalized eigenvalue problem in continuous index form. Here

$$\mathbf{H}(\mathbf{Z}';\mathbf{Z}) = \langle\Phi(\mathbf{Z}')|\, \hat{\rm H}\, |\Phi(\mathbf{Z})\rangle \tag{9}$$

$$\mathbf{N}(\mathbf{Z}';\mathbf{Z}) = \langle\Phi(\mathbf{Z}')|\Phi(\mathbf{Z})\rangle\,. \tag{10}$$

A strength of the GCM representation is that evaluating the matrix elements Eq. (9), (10), is inexpensive, even between single Slater determinants corresponding to different basis sets or when also evaluating operators between such states, by using the Löwdin or Onishi formulae (see Ref. [5] for the combined expression and original references).

In practice, the generalized eigenvalue problem Eq. (8) can be solved by explicit discretization. In this case the matrix formulation is given by

$$\mathbf{H}\, \underline{f} = E\, \mathbf{N}\, \underline{f}\,, \tag{11}$$

*Electronic address: bellygrub@phds.ca

for K sample points $\mathbf{Z}^{(1)}, \ldots, \mathbf{Z}^{(K)}$ and the, to be determined, weightings $f_k = f(\mathbf{Z}^{(k)})$. Using a naive discretization, the computational demand scales exponentially with the dimension D, the number of generator coordinates $\mathbf{Z}$. If one takes modestly $N \approx 10$ points per dimension then the number of samples is $K = N^D = 10^D$, quickly becoming intractable.

Applications in nuclear structure tend to use only a few ($D \leq 3$) generator coordinates, selecting only those operators (such as the quadrupole operator) which correspond to physically motivated nuclear deformation modes in the expansion, rather than the full space of mono-excitation operators. This initial choice was due to the phenomenological origin of the GCM and applications of interest. Subsequent investigations over the last sixty years have been hampered in no small part due to the poor scaling behaviour with the increase in the number of dimensions. This is also true even when the GCM formalism is only used as a means of symmetry projection for even a single broken symmetry Hartree-Fock (HF) or Hartree-Fock-Bogoliubov (HFB) state and the weightings $f(\mathbf{Z})$ are fixed by symmetry. Other approaches have parameterized the weighting function $f(\mathbf{Z})$ by a combination of gaussian functions to restore some of the continuous flavour of the GCM ansatz, however the underlying numerical issues still remain.

To circumvent these difficulties, we propose an alternate formulation of the GCM. This is possible by using a hole-particle representation of the generator coordinate operators which then allows for analytic symmetry projection using tensor operators and the invariant mean (operator average) [9, 10]. Symmetry projection then becomes a "back of an envelope calculation". Explicitly, such mono-excitation generators $\hat{\mathrm{Z}}$ of the hole-particle type are defined as

$$\hat{\mathrm{Z}} = \sum_{AI} Z_{AI}\, \hat{\mathrm{a}}_A^\dagger \hat{\mathrm{a}}_I \,, \tag{12}$$

when generating the Hartree-Fock (HF) class of states $|\Phi(\mathbf{Z})\rangle$ and

$$\begin{aligned} \hat{\mathrm{Z}} = &\sum_{AI} Z_{AI}\, \hat{\mathrm{a}}_A^\dagger \hat{\mathrm{a}}_I \\ &+ \tfrac{1}{2} \sum_{AB} Z^*_{AB}\, \hat{\mathrm{a}}_A^\dagger \hat{\mathrm{a}}_B^\dagger + \tfrac{1}{2} \sum_{IJ} Z_{IJ}\, \hat{\mathrm{a}}_I \hat{\mathrm{a}}_J \,, \end{aligned} \tag{13}$$

when generating the Hartree-Fock-Bogoliubov (HFB) class of states. Here I, J index the occupied spin orbitals (holes) and A, B the unoccupied spin orbitals (particles) with respect to the single-determinantal reference $|\Phi_0\rangle$.

The resulting GCM formulation has a direct connection with projected Hartree-Fock (PHF) and projected Hartree-Fock-Bogoliubov (PHFB) wavefunctions, which then correspond to the single term approximation of the explicitly projected GCM. It is also related to the coupled cluster (CC) approach [11, 12] (see also Refs. [13, 14]). A mapping from the general GCM prescription to CC can be constructed, even when no explicit symmetry breaking is present. This mapping between GCM and CC leads to a distribution-like parameterization of the cluster amplitudes, and an alternate route to solve for the GCM wavefunction via the CC equations. The following sections will develop this new prescription.

II. MAPPING FROM GCM TO CC

In this section a mapping from the GCM representation

$$|\Psi_{\mathrm{GCM}}\rangle = \int d\mathbf{Z}\, f(\mathbf{Z})\, e^{\hat{\mathrm{Z}}} |\Phi_0\rangle \,, \tag{14}$$

to the coupled cluster representation

$$|\Psi_{\mathrm{CC}}\rangle = e^{\hat{\mathrm{T}}} |\Phi_0\rangle \,, \tag{15}$$

of the wavefunction will be elucidated. Here

$$\hat{\mathrm{T}} = \hat{\mathrm{T}}_1 + \hat{\mathrm{T}}_2 + \hat{\mathrm{T}}_3 + \hat{\mathrm{T}}_4 + \ldots \tag{16}$$

are the conventional mono-, bi- and higher order cluster excitation operators (see *e.g.* Refs. [13, 14]). Explicitly,

$$\begin{aligned} \exp(\hat{\mathrm{T}}) = 1 \\ &+ \hat{\mathrm{T}}_1 \\ &+ \hat{\mathrm{T}}_2 + \frac{1}{2}\hat{\mathrm{T}}_1^2 \\ &+ \hat{\mathrm{T}}_3 + \hat{\mathrm{T}}_2\hat{\mathrm{T}}_1 + \frac{1}{3!}\hat{\mathrm{T}}_1^3 \\ &+ \hat{\mathrm{T}}_4 + \hat{\mathrm{T}}_3\hat{\mathrm{T}}_1 + \frac{1}{2!}\hat{\mathrm{T}}_2^2 + \frac{1}{2!}\hat{\mathrm{T}}_2\hat{\mathrm{T}}_1^2 + \frac{1}{4!}\hat{\mathrm{T}}_1^4 + \ldots \,, \end{aligned} \tag{17}$$

where terms corresponding to the same excitation order have been grouped. The mapping from GCM to CC can be expressed from two different perspectives on how the symmetry projection is accomplished: either when the weightings $f(\mathbf{Z})$ are constrained via external means (such as via Lagrange multipliers), or when the weightings are unconstrained but an explicit projection has been performed, which the authors recommended as it is the most computationally efficient. These mappings from GCM to CC are also possible even when symmetry projection is not present or required.

A. Constrained Weighting Formulation

The GCM wavefunction Eq. (14) is generally formulated in terms of broken symmetry generators $\hat{\mathrm{Z}}$ as in Eqs. (12) or (13) however the final wavefunction $|\Psi_{\mathrm{GCM}}\rangle$ is also implicitly assumed to have good quantum numbers and therefore be symmetry adapted. Unless an explicit projection has already been performed (as will be illustrated in the next section), then the weightings $f(\mathbf{Z})$ must satisfy additional constraints to ensure symmetry

is preserved for the overall wavefunction. This can be accomplished by an implicit parameterization (if possible) or by the use of additional constraints imposed via Lagrange multipliers. In either approach, the resulting GCM wavefunction is then given by

$$|\Psi_{\rm GCM}\rangle = \int d\mathbf{Z}\, f_c(\mathbf{Z})\, e^{\hat{\rm Z}}\, |\Phi_0\rangle\,, \tag{18}$$

where $f_c(\mathbf{Z})$ now denote the constrained weights which ensure symmetry is preserved. Possible methods to determine these weights include by explicit incorporation of the projection via a Baker-Campbell-Hausdorff (BCH) expansion (which also effectively modifies $\hat{\rm Z}$) or by requiring the symmetry violating contributions vanish at each order; similar to the Brillouin conditions for HFB. How these weights can be obtained will not however be considered further in this work. Instead a mapping to the coupled cluster form of the wavefunction will now be developed.

The constrained weighting formulation of GCM Eq. (18) can be compared with the coupled cluster representation Eq. (15),

$$e^{\hat{\rm T}}\, |\Phi_0\rangle = \int d\mathbf{Z}\, f_c(\mathbf{Z})\, e^{\hat{\rm Z}}\, |\Phi_0\rangle\,, \tag{19}$$

for each excitation order $\hat{\rm Z}^k$, as compared to the corresponding k-th row in the cluster expression in Eq. (17). At zeroth order,

$$1 = \int d\mathbf{Z}\, f_c(\mathbf{Z})\,, \tag{20}$$

one obtains a normalization condition on the constrained weights $f_c(\mathbf{Z})$. One is tempted to treat $f_c(\mathbf{Z})$ as a distribution, however the scalar weights are themselves generally complex-valued and even when real valued can have regions in the parameter space where the weight is negative.

Turning to the first order term in the expansion,

$$\hat{\rm T}_1 = \int d\mathbf{Z}\, f_c(\mathbf{Z})\, \hat{\rm Z}\,, \tag{21}$$

this equality suggests the interpretation that under the GCM to CC mapping, the coupled cluster mono-excitation operator $\hat{\rm T}_1$ corresponds to an average over the 'pseudo-density' $f_c(\mathbf{Z})$. Explicitly, in terms of the generator coordinate parameters, with $\hat{\rm T}_1$ defined in a similar fashion to Eq. (6),

$$\begin{aligned} T_\mu &= \int dZ_1 \ldots dZ_\mu \ldots dZ_D\, f_c(\mathbf{Z})\, Z_\mu \\ &= \int dZ_\mu\, f_c^{(\mu)}(Z_\mu)\, Z_\mu\,, \end{aligned} \tag{22}$$

where

$$f_c^{(\mu)}(Z_\mu) = \int dZ_1 \ldots dZ_{\mu-1} dZ_{\mu+1} \ldots dZ_D\, f_c(\mathbf{Z})\,, \tag{23}$$

is a reduced (constrained) weight obtained by integrating over all generator coordinates $\mathbf{Z}$ except for Z_μ. From the normalization condition Eq. (20), then

$$\int dZ_\mu\, f_c^{(\mu)}(Z_\mu) = 1\,, \tag{24}$$

for all choices of mono-excitation index μ. This again suggests the interpretation that $f_c(\mathbf{Z})$ can be considered a (complex valued) distribution, in which case $f_c^{(\mu)}(Z_\mu)$ is then a one-parameter reduced weight.

At quadratic ($\hat{\rm Z}^2$) order in the expansion,

$$\left(\hat{\rm T}_2 + \frac{1}{2}\hat{\rm T}_1^2\right) = \frac{1}{2}\int d\mathbf{Z}\, f_c(\mathbf{Z})\, \hat{\rm Z}^2\,. \tag{25}$$

Rewriting

$$\begin{aligned} \hat{\rm T}_2 &= \frac{1}{2}\int d\mathbf{Z}\, f_c(\mathbf{Z})\, \hat{\rm Z}^2 - \frac{1}{2}\hat{\rm T}_1^2 \\ &= \frac{1}{2}\int d\mathbf{Z}\, f_c(\mathbf{Z})\, (\hat{\rm Z} - \hat{\rm T}_1)^2\,, \end{aligned} \tag{26}$$

The bi-excitation operator $\hat{\rm T}_2$ obtained from the constrained weighting GCM to CC mapping is the fluctuation (deviation) from the average. As before, one can formulate a two-parameter reduced weighting $f^{(\mu\nu)}(Z_\mu, Z_\nu)$ for $\mu \neq \nu$.

Continuing in a similar fashion, one finds the higher order excitations

$$\hat{\rm T}_k = \frac{1}{k!}\int d\mathbf{Z}\, f_c(\mathbf{Z})\, (\hat{\rm Z} - \hat{\rm T}_1)^k\,, \tag{27}$$

as the moments (or cumulants) in the space of mono-excitations about the mean given by $\hat{\rm T}_1$. This is the underlying structure of GCM, as cast in the constrained weighting formulation.

B. Explicit Projection Formulation

One difficulty with the approach taken in the previous section is that the constrained weights $f_c(\mathbf{Z})$ require either a specific parametrization or the use of external constraints, increasing the difficulty for a practical solution. This constraint on the weightings can be removed if explicit projection is used. The weights $f(\mathbf{Z})$ are then unconstrained and generally will not have the same values as $f_c(\mathbf{Z})$, although a mapping between these quantities is possible.

Consider the explicit projection,

$$\begin{aligned} \hat{\rm P}\, |\Psi_{\rm GCM}\rangle &= \hat{\rm P}\int d\mathbf{Z}\, f(\mathbf{Z})\, e^{\hat{\rm Z}}\, |\Phi_0\rangle \\ &= \int d\mathbf{Z}\, \left(\hat{\rm P}\, f(\mathbf{Z})\, e^{\hat{\rm Z}}\, |\Phi_0\rangle\right)\,. \end{aligned} \tag{28}$$

If the GCM wavefunction is already symmetry adapted, such as by using constrained weights $f_c(\mathbf{Z})$, then this is

identical to the original expression Eq. (14). If this is not the case, then evaluation of the projection even for a single state,

$$\hat{\mathrm{P}}\,|\Phi(\mathbf{Z})\rangle = \hat{\mathrm{P}}\,e^{\hat{\mathrm{Z}}}\,|\Phi_0\rangle\,, \tag{29}$$

can be extremely computationally demanding or even intractable, depending on the symmetry group $\mathbf{G}$ of the projection operator $\hat{\mathrm{P}}$ and which subgroup $\mathbf{H} \subset \mathbf{G}$ the generators $\hat{\mathrm{Z}}$ transform under [15–17]. Spin angular momentum $\mathbf{G} = \mathbf{SU}(2)$ is the first example of such a 'problem group', where exact state projection methods scale with the size of the state space, i.e. the number of possible Slater determinantal configurations.

If the reference $|\Phi_0\rangle$ is however already symmetry adapted and $\hat{\mathrm{Z}}$ is of the hole-particle type (as we have explicitly chosen), then one can determine an analytical form for the symmetry projection through the use of tensor operators and the invariant mean (operator average) [9, 10]. Instead of the state projection or approximate numerical projection, one employs operator projection

$$\hat{\mathrm{U}}_{\mathrm{sa}} = \mathcal{M}_{\mathbf{G}}(e^{\hat{\mathrm{Z}}})\,, \tag{30}$$

in which case

$$\hat{\mathrm{P}}\,e^{\hat{\mathrm{Z}}}\,|\Phi_0\rangle = \hat{\mathrm{U}}_{\mathrm{sa}}|\Phi_0\rangle\,. \tag{31}$$

Here $\mathcal{M}_{\mathbf{G}}(\cdot)$ is the invariant mean, which for discrete groups $\mathbf{G}$ corresponds to the average

$$\mathcal{M}_{\mathbf{G}}(\hat{\mathrm{A}}) = \frac{1}{|\mathbf{G}|}\sum_{\hat{\mathrm{g}}\in\mathbf{G}} \hat{\mathrm{g}}\,\hat{\mathrm{A}}\,\hat{\mathrm{g}}^{-1}\,. \tag{32}$$

For $\mathbf{SU}(2)$ spin projection, see Ref. [10]. This equality Eq. (31) only holds under certain assumptions on the quantum numbers, for the case of $\mathbf{SU}(2)$ that the reference is a closed-shell singlet $S = 0$, however this method can be extended to non-singlet quantum numbers simply by inclusion of additional tensor operator coupling terms weighted with the appropriate Clebsch-Gordon coefficient. The operator projection Eq. (30) then can be expanded for each order in $\hat{\mathrm{Z}}$, and significantly for each order the symmetry projection is algebraic and exact.

The structure of the symmetry adapted correlation operator $\hat{\mathrm{U}}_{\mathrm{sa}}$ can be formally linked to the coupled cluster expansion, so that

$$\hat{\mathrm{U}}_{\mathrm{sa}} = \exp(\hat{\mathrm{S}})\,, \tag{33}$$

where

$$\hat{\mathrm{S}} = \hat{\mathrm{S}}_1 + \hat{\mathrm{S}}_2 + \hat{\mathrm{S}}_3 + \hat{\mathrm{S}}_4 + \ldots \tag{34}$$

are the symmetry adapted excitations, in correspondence with Eq. (16). Comparing Eqs. (30) with (33), then

$$\hat{\mathrm{S}}_1 = \mathcal{M}_{\mathbf{G}}(\hat{\mathrm{Z}}) \tag{35}$$

$$\hat{\mathrm{S}}_2 = \mathcal{M}_{\mathbf{G}}\left(\frac{1}{2}\hat{\mathrm{Z}}^2\right) - \frac{1}{2}\hat{\mathrm{S}}_1^2 \tag{36}$$

$$\hat{\mathrm{S}}_3 = \mathcal{M}_{\mathbf{G}}\left(\frac{1}{3!}\hat{\mathrm{Z}}^3\right) - \hat{\mathrm{S}}_2\hat{\mathrm{S}}_1 - \frac{1}{3!}\hat{\mathrm{S}}_1^3 \tag{37}$$

$$\hat{\mathrm{S}}_4 = \mathcal{M}_{\mathbf{G}}\left(\frac{1}{4!}\hat{\mathrm{Z}}^4\right) - \hat{\mathrm{S}}_3\hat{\mathrm{S}}_1 - \frac{1}{2}\hat{\mathrm{S}}_2^2 - \frac{1}{2}\hat{\mathrm{S}}_2\hat{\mathrm{S}}_1^2 - \frac{1}{4!}\hat{\mathrm{S}}_1^4\,, \tag{38}$$

and so on. This approach was advocated in our previous work [10] as an exact method at any order to analytically determine projected broken symmetry wavefunctions, including for higher order operators approximations (*e.g.* PUCCSD). Once the symmetry group $\mathbf{G}$ is known, explicit forms for the invariant mean $\mathcal{M}_{\mathbf{G}}$ are relatively straightforward to evaluate for any k-body operator, particularly when $\hat{\mathrm{Z}}$ is represented in terms of tensor operators.

As a simple example of this symmetry projection, consider the generator $\hat{\mathrm{Z}}$ corresponding to real-valued BCS states,

$$\begin{aligned}\hat{\mathrm{Z}} &= \sum_{N,S,M} \hat{\mathrm{Z}}^{(N,S,M)} \\ &= \sum_{N=-1}^{+1} \hat{\mathrm{Z}}_R^{(N,0,0)}\,,\end{aligned} \tag{39}$$

given in terms of its tensor components $\hat{\mathrm{Z}}^{(N,S,M)}$. The corresponding cluster operators $\hat{\mathrm{S}}$ are given by

$$\hat{\mathrm{S}}_1 = \hat{\mathrm{Z}}_R^{(0,0,0)} \tag{40}$$

$$\hat{\mathrm{S}}_2 = \hat{\mathrm{Z}}_R^{(1,0,0)}\,\hat{\mathrm{Z}}_R^{(-1,0,0)} \tag{41}$$

$$\hat{\mathrm{S}}_3 = 0 \tag{42}$$

$$\begin{aligned}\hat{\mathrm{S}}_4 &= -\frac{1}{4}\left(\hat{\mathrm{Z}}_R^{(1,0,0)}\right)^2\left(\hat{\mathrm{Z}}_R^{(-1,0,0)}\right)^2 \\ &= -\frac{1}{4}\,\hat{\mathrm{S}}_2^2\,,\end{aligned} \tag{43}$$

which can be verified via Eqs. (35)–(38). This can similarly be applied to other symmetry groups. Explicit expressions for $\mathcal{M}_{\mathbf{G}}(\cdot)$ operator projection of up to fourth order will be provided in a forthcoming paper [18] for the symmetry group $\mathbf{G} = \mathbf{S} \times \mathbf{T} \times \mathbf{N}$, which corresponds to the projected real and complex-valued RHF, UHF, GHF, BCS and HFB classes of wavefunctions.

For the remainder of this paper it is taken that the assumptions required for the invariant mean projection Eq. (31) to be valid hold. For specificity, in case of the quantum chemistry literature this would require that the orbitals are real valued canonical spin-orbitals (space × spin) and that the reference $|\Phi_0\rangle$ is a closed shell singlet state with good particle number. For simplicity, the generator coordinates $\hat{\mathrm{Z}}$ and weightings $f(\mathbf{Z})$ will also be

assumed real valued. These conditions are intended to simplify the derivation of this alternate GCM prescription, avoiding the potential quagmire with spin coupling and other issues for which a unique definition of the hole-particle representation is not as straightforward. Some of these conditions can be relaxed and the GCM structural form generalized further, such as by allowing for different reference states or by choosing $|\Phi(\mathbf{Z})\rangle$ to include both single and double excitations for the generator coordinates. The authors however wish to first grab the proverbial low-lying fruit first.

Using the operator average, then

$$\hat{\mathrm{P}}|\Psi_{\mathrm{GCM}}\rangle = \hat{\mathrm{P}} \int d\mathbf{Z}\, f(\mathbf{Z})\, \mathcal{M}_{\mathbf{G}}(e^{\hat{Z}})\, |\Phi_0\rangle\,, \tag{44}$$

or using Eqs. (31), (33),

$$\hat{\mathrm{P}}|\Psi_{\mathrm{GCM}}\rangle = \int d\mathbf{Z}\, f(\mathbf{Z})\, e^{\hat{\mathrm{S}}(\hat{\mathrm{Z}})}\, |\Phi_0\rangle\,. \tag{45}$$

In this representation, the GCM is a (continuous) linear combination of cluster operators, each corresponding to a projected broken symmetry wavefunction.

As before, comparing order by order with the coupled cluster expansion

$$1 = \int d\mathbf{Z}\, f(\mathbf{Z})\,, \tag{46}$$

but now for the unconstrained weightings. The mono-excitation operator is given by

$$\begin{aligned}\hat{\mathrm{T}}_1 &= \int d\mathbf{Z}\, f(\mathbf{Z})\, \mathcal{M}_{\mathbf{G}}(\hat{\mathrm{Z}}) \\ &= \int d\mathbf{Z}\, f(\mathbf{Z})\, \hat{\mathrm{S}}_1(\hat{\mathrm{Z}})\,,\end{aligned} \tag{47}$$

and so the unconstrained mono-excitation $\hat{\mathrm{T}}_1$ is an average over projected mono-excitation generator $\hat{\mathrm{Z}}$.

Turning to the bi-excitations,

$$\hat{\mathrm{T}}_2 + \frac{1}{2}\hat{\mathrm{T}}_1^2 = \frac{1}{2}\int d\mathbf{Z}\, f(\mathbf{Z})\, \mathcal{M}_{\mathbf{G}}(\hat{\mathrm{Z}}^2)\,, \tag{48}$$

or

$$\hat{\mathrm{T}}_2 = \frac{1}{2!}\int d\mathbf{Z}\, f(\mathbf{Z})\, \mathcal{M}_{\mathbf{G}}(\hat{\mathrm{Z}}^2) - \frac{1}{2!}\hat{\mathrm{T}}_1^2\,. \tag{49}$$

The bi-excitation can be interpreted, just as before, as the deviation from the average amplitude

$$\hat{\mathrm{T}}_2 = \frac{1}{2!}\int d\mathbf{Z}\, f(\mathbf{Z})\, \mathcal{M}_{\mathbf{G}}((\hat{\mathrm{Z}} - \hat{\mathrm{T}}_1)^2)\,, \tag{50}$$

where however the symmetry projection via the invariant mean $\mathcal{M}_{\mathbf{G}}(\cdot)$ has been incorporated with unconstrained weightings $f(\mathbf{Z})$.

Alternately, the bi-excitations can be re-cast using the cluster expansion for each term,

$$\begin{aligned}\hat{\mathrm{T}}_2 &= \int d\mathbf{Z}\, f(\mathbf{Z})\, \left(\frac{1}{2}\hat{\mathrm{S}}_1^2(\hat{\mathrm{Z}}) + \hat{\mathrm{S}}_2(\hat{\mathrm{Z}})\right) - \frac{1}{2!}\hat{\mathrm{T}}_1^2 \\ &= \int d\mathbf{Z}\, f(\mathbf{Z})\, \hat{\mathrm{S}}_2(\hat{\mathrm{Z}}) \\ &\quad + \frac{1}{2}\int d\mathbf{Z}\, f(\mathbf{Z})\, \left(\hat{\mathrm{S}}_1(\hat{\mathrm{Z}}) - \hat{\mathrm{T}}_1\right)^2\,,\end{aligned} \tag{51}$$

in which case the $\hat{\mathrm{T}}_2$ comes both from the sum of a weighted $\hat{\mathrm{S}}_2$ and the deviation from the operator average of $\hat{\mathrm{T}}_1$. Here the $\hat{\mathrm{S}}_2$ contribution comes from explicit symmetry breaking, whereas the second term comes from the spread of the mono-excitation coefficients or alternately the cross-correlation. The $\hat{\mathrm{S}}_2$ term vanishes entirely if no symmetry breaking is present and can be interpreted as the canonical order parameter. One finds that $\hat{\mathrm{T}}_2$ only vanishes if both $\hat{\mathrm{S}}_2 = 0$ and

$$f(\mathbf{Z}) = \delta(\mathbf{Z} - \mathbf{Z}_0)\,, \tag{52}$$

that is that the GCM corresponds to a single symmetry restricted Slater determinant, *i.e.* restricted Hartree-Fock (RHF) wavefunction.

At the $\hat{\mathrm{T}}_2$ bi-excitation level, GCM is a (continuous) linear combination of projected broken symmetry wavefunctions as in Eq. (45) or instead via Eq. (51) is a weighted distribution of pairing amplitudes. The latter scheme can be related to the plethora of approximate doubles schemes, such as perfect pairing, RVB, GVB, canonical BCS, AGP, and so on. This representation thus provides a natural framework to discuss the sparsity structure of the cluster amplitudes, which can be dictated by symmetry or other considerations.

For the third order expression,

$$\hat{\mathrm{T}}_3 + \hat{\mathrm{T}}_2\hat{\mathrm{T}}_1 + \frac{1}{3!}\hat{\mathrm{T}}_1^3 = \frac{1}{3!}\int d\mathbf{Z}\, f(\mathbf{Z})\mathcal{M}_{\mathbf{G}}(\hat{\mathrm{Z}}^3)\,. \tag{53}$$

Re-arranging,

$$\hat{\mathrm{T}}_3 = \frac{1}{3!}\int d\mathbf{Z}\, f(\mathbf{Z})\, \mathcal{M}_{\mathbf{G}}((\hat{\mathrm{Z}} - \hat{\mathrm{T}}_1)^3)\,, \tag{54}$$

which can be linked directly to the corresponding symmetry constrained weighting $f_c(\mathbf{Z})$ expression. Alternately, in terms of the symmetry adapted cluster operators,

$$\begin{aligned}\hat{\mathrm{T}}_3 &= \int d\mathbf{Z}\, f(\mathbf{Z})\, \hat{\mathrm{S}}_3(\mathbf{Z}) \\ &\quad + \int d\mathbf{Z}\, f(\mathbf{Z})\, (\hat{\mathrm{S}}_2(\mathbf{Z}) - \hat{\mathrm{T}}_2)(\hat{\mathrm{S}}_1(\mathbf{Z}) - \hat{\mathrm{T}}_1) \\ &\quad + \frac{1}{3!}\int d\mathbf{Z}\, f(\mathbf{Z})\, (\hat{\mathrm{S}}_1(\mathbf{Z}) - \hat{\mathrm{T}}_1)^3\,.\end{aligned} \tag{55}$$

The expansion suggests that the higher order cluster amplitudes, when interpreted through the GCM to CC mapping, are a combination of 'pure' symmetry breaking contributions together with distributional (cross-correlation) contributions.

Fourth order proceeds similarly,

$$\hat{\mathrm{T}}_4 = \frac{1}{4!}\int d\mathbf{Z}\, f(\mathbf{Z})\, \mathcal{M}_{\mathbf{G}}((\hat{\mathrm{Z}} - \hat{\mathrm{T}}_1)^4)\,, \tag{56}$$

and for the cluster formulation

$$\begin{aligned}\hat{\mathrm{T}}_4 = &\int d\mathbf{Z}\, f(\mathbf{Z})\, \hat{\mathrm{S}}_4(\mathbf{Z}) \\ &+ \int d\mathbf{Z}\, f(\mathbf{Z})\, (\hat{\mathrm{S}}_3(\mathbf{Z}) - \hat{\mathrm{T}}_3)(\hat{\mathrm{S}}_1(\mathbf{Z}) - \hat{\mathrm{T}}_1) \\ &+ \frac{1}{2!}\int d\mathbf{Z}\, f(\mathbf{Z})\, (\hat{\mathrm{S}}_2(\mathbf{Z}) - \hat{\mathrm{T}}_2)^2 \\ &+ \frac{1}{2!}\int d\mathbf{Z}\, f(\mathbf{Z})\, (\hat{\mathrm{S}}_2(\mathbf{Z}) - \hat{\mathrm{T}}_2)\, (\hat{\mathrm{S}}_1(\mathbf{Z}) - \hat{\mathrm{T}}_1)^2 \\ &+ \frac{1}{4!}\int d\mathbf{Z}\, f(\mathbf{Z})\, (\hat{\mathrm{S}}_1(\mathbf{Z}) - \hat{\mathrm{T}}_1)^4 \end{aligned} \tag{57}$$

This is the structure of the GCM under the explicitly projected CC mapping, which provides an alternate route to a highly efficient and direct means of symmetry projection.

III. FIRST CHECKPOINT

This work has provided a clear prescription for how to utilize the structural form of GCM while avoiding the historical computational bottleneck. This has been accomplished through the use of analytical projection via the invariant mean and tensor operator representations, which can be effected efficiently when the generator coordinate is cast within the hole-particle representation.

At the lowest level of approximation, which takes a single Slater determinant for the GCM, this approach yields an efficient method to obtain the wavefunction for individual PHF and PHFB states as cast within the coupled cluster representation. The imposed sparsity on the cluster amplitudes (for a single or linear combination of Slater determinants) can be formulated in this framework, and related to the canonical (finite size) order parameters of the system. For higher order amplitudes, the symmetry projection can also be used to obtain the externally corrected correlation corrections for the $\hat{\mathrm{T}}_3$ and $\hat{\mathrm{T}}_4$ amplitudes. These can then be determined self-consistently through the higher order cluster equations, as significantly fewer free parameters are required as than a full $\hat{\mathrm{T}}_3$ or $\hat{\mathrm{T}}_4$ calculation.

The GCM expansion provides a natural approach to go beyond projected HF and projected HFB wavefunctions towards the exact solution, and in this work has been mapped to the coupled cluster amplitudes. Under this mapping, the resulting GCM weights $f(\mathbf{Z})$ can be interpreted as a pseudo-distribution. Provided the weights $f(\mathbf{Z})$ between uncoupled subsystems are uncorrelated, then the GCM wavefunction is size-extensive, just as for the coupled cluster approach.

An explicit solution procedure has not been presented in this work, however one can use the coupled cluster equations directly as the authors have advocated previously for projected broken symmetry wavefunctions [10]. Even at the single term approximation of GCM, which corresponds to the PHF or PHFB wavefunctions, this leads to a reduction of the cluster equations to a relatively few expressions for the free parameters. This approach can also be used for GCM even when symmetry breaking is not present, such as when choosing D physically motivated generator coordinates. The coupled cluster equations then give a D^6 scaling, once intermediates have been calculated.

The mapping from GCM to CC provides one noteworthy point for the coupled cluster approach, illustrating the severity of conventional coupled cluster truncation schemes. In the case of CCSD, singles and doubles amplitudes $\hat{\mathrm{T}}_1$ and $\hat{\mathrm{T}}_2$ are evaluated but all higher order amplitudes $\hat{\mathrm{T}}_k$ for $k > 2$ are omitted (zeroed). This places a strong constraint on the pseudo-distribution $f(\mathbf{Z})$ which results in unphysical artifacts in the resulting self-consistent CCSD equations. One avenue for further investigation is how the pseudo-distributional structures for $\hat{\mathrm{T}}_3$ and $\hat{\mathrm{T}}_4$ can be exploited to address this deficiency.

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